

Article principal
recouvrant en partie

la T H E S E présentée
pour l'obtention
du grade de DOCTEUR ES SCIENCES
PHYSIQUES
A L'UNIVERSITE DE PARIS VI

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Sujet de la thèse : Quelques comparaisons de traitements semi-classiques et quantiques de l'interaction entre le champ électromagnétique et des systèmes quantiques à spectre discret.

soutenue le : 14 JUIN 1974

devant la Commission d'examen composée de :

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Spontaneous-Emission Feedback in a Three-Level Quantum System: A Case of Conflict between Semi-Classical and Quantum Theories of Radiation.

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(ricevuto il 26 Giugno 1972)

Summary. — The behaviour of a three-level quantum system with a fundamental state and two closely grouped excited states is studied during its interaction with a strong monochromatic electromagnetic field. The EMF couples strongly and in a resonant manner the fundamental level (first level) with one excited level (second level) and couples weakly and in a nonresonant manner the fundamental level with the other excited one (third level). The semi-classical theory of radiation of Jaynes and Crisp and the quantum theory of radiation, in Lehmberg's formalism, are successively used to calculate the density matrix of the system. It is found that, for given parameters characterizing the electromagnetic field and the first two levels, there are two particular values of the frequency gap between the excited levels for which the third level can be strongly populated. For the first value the two theories give consistent results and the third level is populated under both the effects of the external field and the field spontaneously emitted by the system (reactant or internal field). For the second value, only the semi-classical theory predicts a strong population for the third level and it is populated under the effect of the internal field alone. The values of the frequency gap are approximately predicted by using the concepts of « pseudolevels » in the semi-classical case and of « semi-dressed » system in the quantum case. The predictions are verified and improved by numerical calculations. It is proposed to use the discrepancy between the two theories in the second case to test experimentally the validities of the semi-classical and quantum theories of radiation.

1. — Introduction.

For some years several authors ⁽¹⁾ have attempted to build a semi-classical theory of the interaction between light and matter which, they claim, can ac-

⁽¹⁾ See ref. ⁽²⁾ for a comprehensive bibliography.

⁽²⁾ J. F. CLAUSER: *Phys. Rev. A*, **6**, 49 (1972).

count for many phenomena as satisfactorily as the quantum theory of radiation. JAYNES *et al.* ^(3,5) and NESBET ^(6,7) have particularly investigated the spontaneous emission of light, by a quantum system with and without an external electromagnetic field. As in the quantum theory an excited quantum system can emit light spontaneously but its general behaviour is different in the semi-classical and quantum approaches. Most experiments cannot discriminate at the moment between the results of JAYNES *et al.* and those of quantum theory. On the contrary, NESBET ⁽⁷⁾ has found one case (Raman scattering) where the semi-classical theory fails to explain the intensity of the anti-Stokes transition. More recently CLAUSER ⁽²⁾ has shown that the semi-classical theories do not agree with the photon correlation experiment of KOCHER and COMMINS ⁽⁸⁾, which is correctly explained by quantum theory. In this paper we show that in a three-level quantum system interacting with a strong monochromatic electromagnetic field and under certain conditions the spontaneous emission from one of the excited states reacts upon the system and is responsible for a strong population of the second excited state. If we make the same calculation using quantum theory of radiation, we find that, for the same physical conditions, no anomalous population of the second excited state is predicted. There exist other sets of parameters for which an anomalous population of the second excited state is predicted by the two theories but then the phenomenon arises from the interaction with both the external field and the reactant field. But, as for the spontaneous decay rate, the behaviour of the quantum system is not exactly the same in the two theories. In the case where only the semi-classical theory predicts a strong population of the second excited level an «all-or-nothing» experiment may decide which of the two theories must be regarded with some suspicion when calculating the effect of light on matter.

2. - Semi-classical treatment.

We consider a quantum system with levels $|1\rangle$, $|2\rangle$ and $|3\rangle$ of energies 0, $\hbar\Omega_2$, $\hbar\Omega_3$. The dipolar electric moments are defined by $\mu_{12} = \langle 1|\mu|2\rangle$ and $\mu_{13} = \langle 1|\mu|3\rangle$. We assume $\mu_{23} = 0$. φ is the angle between μ_{12} and μ_{13} . The system interacts with an electromagnetic field $E(t) = E_0 \cos \Omega t$. The other parameters which define the interaction are $\delta = \Omega_2 - \Omega$, $\Delta = \Omega_3 - \Omega_2$, $V_{12} = (1/2\hbar)E_0 \cdot \mu_{12}$, $V_{13} = (1/2\hbar)E_0 \cdot \mu_{13}$. It is assumed that $|V_{12}| \gg |V_{13}|$, $|V_{13}| \ll \delta + \Delta$ and $|V_{12}| \geq \delta$. To use the system of differential equations, given by

⁽³⁾ E. T. JAYNES and F. W. CUMMINGS: *Proc. IEEE*, **51**, 89 (1963).

⁽⁴⁾ M. D. CRISP and E. T. JAYNES: *Phys. Rev.*, **179**, 1253 (1969).

⁽⁵⁾ C. R. STROUD and E. T. JAYNES: *Phys. Rev. A*, **1**, 106 (1970).

⁽⁶⁾ R. K. NESBET: *Phys. Rev. A*, **4**, 259 (1971).

⁽⁷⁾ R. K. NESBET: *Phys. Rev. Lett.*, **27**, 553 (1971).

⁽⁸⁾ C. A. KOCHER and E. D. COMMINS: *Phys. Rev. Lett.*, **18**, 575 (1967).



JAYNES and CRISP (⁴), which governs the time evolution of the density matrix of the quantum system in the interaction picture we also need the quantities

$$(1) \quad A_{ij}^{\beta\alpha} = \frac{4}{3} \mu_{\alpha\beta} \cdot \mu_{ij} (\Omega_\alpha - \Omega_\beta)^2 (\Omega_i - \Omega_j) / \hbar c^3.$$

In the equations (A2) of JAYNES and CRISP (⁴) any term of the form $A_{ij}^{\beta\alpha} \varrho_{\beta\alpha} \exp [i(\Omega_\alpha - \Omega_\beta)t]$ is the matrix element between eigenstates $\langle i|$ and $|j\rangle$ of the operator which represents the interaction between the dipolar moment and the internal electromagnetic field corresponding to the polarization $\mu_{\alpha\beta}(t)$. The polarization $\mu_{\alpha\beta}(t)$ is proportional to $\varrho_{\beta\alpha}(t)$. The frequencies of the internal field are then the frequencies of any element $\varrho_{kl}(t)$ of the density matrix, if there exists also a nonzero dipolar moment μ_{lk} . We also assume $|V_{12}| \ll \Omega$, so that the rotating-wave approximation can be used. This last hypothesis is valid to a very good approximation for a near resonant interaction in the optical frequency range and with the light sources available now. Under these conditions the frequencies of the polarization lie in a frequency range centred around Ω and of half-width of the order of $2V_{12} + |A|$. The internal field is weak but in some cases it can induce resonant transitions between the fundamental and the excited states. Our purpose is to find under which conditions the internal field can strongly populate an excited level of the system, which cannot be appreciably populated by the interaction with the external field. The differential equations of JAYNES and CRISP (⁴) cannot be analytically solved. So it is very useful to have a method which qualitatively predicts the resonance condition for a given transition under the effect of the internal field. This can be conveniently performed by using the concept of *pseudolevels*. To explain this concept and its use by an example we revert to the traditional semi-classical theory of interaction, without the reaction of the matter on the field, with the same quantum system. We have treated this problem in another paper (⁹) (referred to as paper I). The wave function of the system during the interaction can be written

$$(2) \quad \psi(t) = \sum_{i=1}^3 c_i(t) |i\rangle \exp [-i\Omega_i t],$$

where

$$(3) \quad c_i(t) = \sum_{k=1}^3 a_{ik} \exp [i\lambda_{i,k} t].$$

Equation (2) can be rewritten

$$(4) \quad \psi(t) = \sum_{i,k} a_{ik} \exp [-i(\Omega_i - \lambda_{i,k})t] |i\rangle.$$

(⁹) G. OLIVER: *Physica*, **51**, 197 (1971). Referred to as paper I.

AUTLER and TOWNES ⁽¹⁰⁾ have introduced the idea that each term $|i\rangle \exp[-i \cdot (\Omega_i - \lambda_{i,k})t]$ can be considered as a component of a so-called « spectrum ». Each component has the appearance of a level but it is not a stationary eigenstate of a Hamiltonian. However this concept is very useful because, when one adds a weak interaction to the Hamiltonian, it will be resonant if it has the same frequency as the transition between two components of the « spectrum ». We call such a term as $\exp[-i(\Omega_i - \lambda_{i,k})t]|i\rangle$ the wave function of a *pseudolevel* and we say that $\hbar(\Omega_i - \lambda_{i,k})$ is its *pseudoenergy*. In paper I the pseudolevel concept has been used to explain the anomalous population of level $|3\rangle$ under the effect of an external field nearly resonant with the transition 1-2.

To have simple expressions we let $\delta = 0$. If only the interaction V_{12} is taken into account the « spectrum » is made up with a true level $|3\rangle$ and four pseudolevels

$$(5a) \quad |I\rangle = |1\rangle \exp[iV_{12}t],$$

$$(5b) \quad |II\rangle = |1\rangle \exp[-iV_{12}t],$$

$$(5c) \quad |III\rangle = |2\rangle \exp[-i(\Omega_2 - V_{12})t],$$

$$(5d) \quad |IV\rangle = |2\rangle \exp[-i(\Omega_2 + V_{12})t],$$

with pseudoenergies

$$(6a) \quad E_I = -\hbar V_{12},$$

$$(6b) \quad E_{II} = \hbar V_{12},$$

$$(6c) \quad E_{III} = \hbar(\Omega_2 - V_{12}),$$

$$(6d) \quad E_{IV} = \hbar(\Omega_2 + V_{12}).$$

If we now introduce the interaction V_{13} , it couples the level $|3\rangle$ with the pseudolevels $|I\rangle$ and $|II\rangle$. As the frequency of the external field is Ω_2 and $V_{13} \ll 2V_{12}$, it is obvious that the transition between $|II\rangle$ and $|3\rangle$ will be resonant if $\Delta \approx V_{12}$ and then the level $|3\rangle$ will be strongly populated. The exact calculation agrees with these predictions very well. We have shown in another paper ⁽¹¹⁾ (referred to as paper II) that the $c_i(t)$'s in $\psi(t)$ are always sums of imaginary exponentials whatever the number of levels and the number of frequencies in the electromagnetic field are.

In our problem the pseudolevel method does not give us an exact solution because the internal field is itself depending on the pseudoenergies of the pseudolevels. It only allows us to make a qualitative prediction but it is yet very

⁽¹⁰⁾ S. H. AUTLER and C. H. TOWNES: *Phys. Rev.*, **100**, 703 (1955).

⁽¹¹⁾ G. OLIVER: *Lett. Nuovo Cimento*, **2**, 1075 (1971). Referred to as paper II.

useful. First consider the interaction V_{12} . The spectrum is again made with four pseudolevels and a level. Without the restriction $\delta = 0$, the pseudolevels and their energies can be written

$$(7a) \quad |I\rangle = |1\rangle \exp[-i\Omega_I t],$$

$$(7b) \quad |II\rangle = |1\rangle \exp[-i\Omega_{II} t],$$

$$(7c) \quad |III\rangle = |2\rangle \exp[-i\Omega_{III} t],$$

$$(7d) \quad |IV\rangle = |2\rangle \exp[-i\Omega_{IV} t],$$

where

$$(8a) \quad \Omega_I = \frac{1}{2}\delta - \beta,$$

$$(8b) \quad \Omega_{II} = \frac{1}{2}\delta + \beta,$$

$$(8c) \quad \Omega_{III} = \Omega + \frac{1}{2}\delta - \beta,$$

$$(8d) \quad \Omega_{IV} = \Omega + \frac{1}{2}\delta + \beta$$

with

$$(8e) \quad \beta = \frac{1}{2}(\delta^2 + 4V_{12}^2)^{\frac{1}{2}}.$$

Now we add the weak interaction with the external field V_{13} , and the interaction with the internal field. The internal field is proportional to the time derivative of the polarization $\mathbf{p}_{12}(t) = \boldsymbol{\mu}_{12}(t) + \boldsymbol{\mu}_{21}(t)$. It has three components at frequencies $\Omega - 2\beta$, Ω and $\Omega + 2\beta$. But if $\delta = 0$, the weight of the component at frequency Ω is zero. In the rotating-wave approximation the interaction matrix elements which govern, in the differential equations of JAYNES and CRISP (⁴), the interaction between the system and the internal field corresponding to the polarization $\mathbf{p}_{12}(t)$ are $\frac{1}{2}A_{12}^{12}$, $\frac{1}{2}A_{21}^{21}$, $\frac{1}{2}A_{13}^{12}$ and $\frac{1}{2}A_{31}^{21}$. The absolute value of the first two is $\frac{1}{2}A_{21}$, where A_{21} is the Einstein coefficient corresponding to the spontaneous emission from state $|2\rangle$. (Do not confuse the four-subscript A with the two-subscript Einstein coefficients.) As we have assumed $\Delta \ll \Omega$, the absolute value of the last two terms can be approximately written $(A_{21}A_{31})^{\frac{1}{2}} \cos \varphi$. As all the results remain unchanged if the signs of δ and Δ are simultaneously changed, only the configuration with $\Delta > 0$ will be considered. This hypothesis implies no physical restriction. From equalities (7) and (8) we can conclude that the components of the field at frequencies Ω and $\Omega + 2\beta$ (external field plus a part of the internal field) will permit a strong population of level $|3\rangle$ if $\Delta \approx \beta - \delta/2$ and the component at frequency $\Omega + 2\beta$ (the internal field alone) will produce a strong population of level $|3\rangle$ if $\Delta \approx 3\beta - \delta/2$. Reciprocally the value of δ (*i.e.* the detuning frequency between the external-field frequency and the frequency of the trans-

ition 1-2) corresponding to a strong population of level $|3\rangle$ shifted from $|2\rangle$ by Δ is

$$(9) \quad \delta = (V_{12}^2 - \Delta^2)/\Delta$$

for the field at frequency Ω , and

$$(10) \quad \delta = \frac{1}{4}(\Delta \pm 3(\Delta^2 - 8V_{12}^2)^{\frac{1}{2}})$$

for the field at frequency $\Omega + 2\beta$. It appears from formula (10) that there are two detuning values for each position of the third level and that the reactant field cannot operate if $\Delta < 2\sqrt{2}V_{12}$. Figure 1 shows the level and pseudo-level scheme of a system where condition (10) with a minus sign is satisfied.

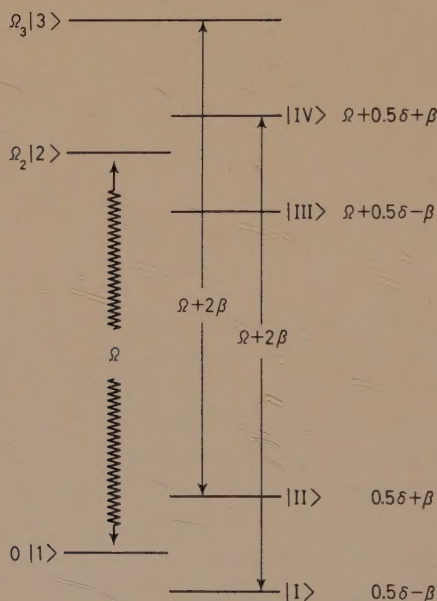


Fig. 1. - Scheme of the pseudolevels of a three-level quantum system when condition (10) is satisfied.

In this paper we are particularly interested in the effect of the reactant field on the quantum system. So we shall study more precisely the behaviour of the system when relation (10) is nearly satisfied. Relation (10) is only a qualitative prediction. In fact, when writing the resonance condition (10) four approximations have been made: 1) We have neglected the nonresonant interaction between pseudolevel $|I\rangle$ and level $|3\rangle$ and the nonresonant interaction between pseudolevel $|II\rangle$ and level $|3\rangle$ due to the two lower-frequency components of the field, 2) the effect of the reactant field corresponding to

the polarization $\mathbf{p}_{12}(t)$ on the 1-2 transition has been neglected, 3) we have also neglected all the effects of the reactant field corresponding to the polarization $\mathbf{p}_{13}(t) = \mu_{13}(t) + \mu_{31}(t)$ on the system, 4) it has been assumed that $\varrho_{12}(t)$ is always the same as if level $|3\rangle$ were not coupled with level $|1\rangle$. Among these four approximations the first and the third are the less drastic. Without approximation 1) the value of δ corresponding to the resonance would be slightly displaced. We have made no exact calculations because approximations 2) and 4) may alter our prediction much more seriously. Numerical calculations have shown that the error on Δ for $\delta = 0$ and a ratio $V_{12}/V_{13} = 10$ is of the order of 0.2 percent. The third approximation is the less restrictive as the polarization $\mathbf{p}_{13}(t)$ has nearly the same frequencies as $\mathbf{p}_{12}(t)$ and is much weaker. The second and fourth approximations mean that we have assumed that the amplitudes and frequencies of the reactant field were constant. In fact the amplitudes cannot be constant firstly because the level $|1\rangle$ is depopulated for the benefit of level $|3\rangle$ and the amplitude of $\mathbf{p}_{12}$ is reduced and secondly because, if $\delta \neq 0$, the internal field corresponding to $\mathbf{p}_{12}(t)$ induces the destruction of the components of $\mathbf{p}_{12}(t)$ with frequencies $\Omega \pm 2\beta$ and the amplitude of the reactant field at frequency $\Omega + 2\beta$ vanishes⁽⁵⁾. On the other hand the frequency of the nutation between states $|1\rangle$ and $|2\rangle$ is slightly modified by the interaction between levels $|1\rangle$ and $|3\rangle$. This modification is itself dependent on the amplitude of the internal field, so that the frequency of the internal field is slightly time-modulated. Therefore the frequency of the internal field is not exactly known. If we assume that $\varrho_{12}(t)$ has constant amplitudes and frequencies during a short time, the time evolution of $\varrho_{33}(t)$ is governed by the four following equations which are the equations (A2) of JAYNES and CRISP⁽⁴⁾ simplified by using the rotating-wave approximation and neglecting all frequency shifts:

$$\begin{aligned}
 (11a) \quad \dot{\varrho}_{11}(t) = & V_{12} \exp[-i\delta t] \varrho_{21} + V_{12} \exp[i\delta t] \varrho_{12} + \\
 & + V_{13} \exp[-i(\delta + \Delta)t] \varrho_{31} + V_{13} \exp[i(\delta + \Delta)t] \varrho_{13} - \\
 & - \frac{1}{2} A_{12}^{12} \varrho_{12} \varrho_{21} - \frac{1}{2} A_{13}^{13} \varrho_{13} \varrho_{21} \exp[i\Delta t] - \frac{1}{2} A_{13}^{12} \varrho_{31} \exp[-i\Delta t] \varrho_{12} - \\
 & - \frac{1}{2} A_{13}^{13} \varrho_{13} \varrho_{31} + \frac{1}{2} A_{21}^{21} \varrho_{21} \varrho_{12} + \frac{1}{2} A_{21}^{31} \varrho_{31} \varrho_{12} \exp[-i\Delta t] + \\
 & + \frac{1}{2} A_{31}^{21} \varrho_{21} \varrho_{13} \exp[i\Delta t] + \frac{1}{2} A_{31}^{31} \varrho_{31} \varrho_{13},
 \end{aligned}$$

$$\begin{aligned}
 (11b) \quad \dot{\varrho}_{33}(t) = & -V_{13} \varrho_{31} \exp[-i(\delta + \Delta)t] - V_{13} \varrho_{13} \exp[i(\delta + \Delta)t] - \\
 & - \frac{1}{2} A_{31}^{31} \varrho_{31} \varrho_{13} - \frac{1}{2} A_{31}^{21} \varrho_{21} \varrho_{13} \exp[i\Delta t] + \frac{1}{2} A_{13}^{13} \varrho_{13} \varrho_{31} + \frac{1}{2} A_{13}^{12} \varrho_{12} \varrho_{31} \exp[-i\Delta t],
 \end{aligned}$$

$$\begin{aligned}
 (11c) \quad \dot{\varrho}_{13}(t) = & +V_{12} \varrho_{23} \exp[-i\delta t] + V_{13} \varrho_{33} \exp[-i(\delta + \Delta)t] - \\
 & - V_{13} \varrho_{11} \exp[-i(\delta + \Delta)t] - \frac{1}{2} A_{12}^{12} \varrho_{12} \varrho_{23} - \frac{1}{2} A_{12}^{13} \varrho_{13} \varrho_{23} \exp[i\Delta t] - \\
 & - \frac{1}{2} A_{13}^{12} \varrho_{12} \varrho_{33} \exp[-i\Delta t] - \frac{1}{2} A_{13}^{13} \varrho_{13} \varrho_{33} + \frac{1}{2} A_{13}^{12} \varrho_{12} \varrho_{11} \exp[-i\Delta t] + \frac{1}{2} A_{13}^{13} \varrho_{13} \varrho_{11},
 \end{aligned}$$

$$(11d) \quad \dot{\varrho}_{31}(t) = \dot{\varrho}_{13}^*(t).$$

One can easily see from these equations that, when condition (10) is satisfied, the resonant interaction between the internal field and the system is governed by the interaction matrix elements $\frac{1}{2} A_{13}^{12} \varrho_{12}(t)$, $\frac{1}{2} A_{13}^{21} \varrho_{21}(t)$, $\frac{1}{2} A_{31}^{12} \varrho_{12}(t)$ and $\frac{1}{2} A_{31}^{21} \varrho_{21}(t)$. As $|\varrho_{12}| \leq \frac{1}{2}$ and only at most one half of the reactant field corresponding to ϱ_{12} is responsible for the resonant interaction, the frequency of the nutation between levels $|1\rangle$ and $|3\rangle$ under the effect of the reactant field is

$$(12) \quad \nu_{13} \leq \frac{1}{4} A_{31}^{12}.$$

This implies $\nu_{13} \ll V_{12}$. We call ν_{12} the nutation frequency between levels $|1\rangle$ and $|2\rangle$ when taking into account all the interactions between the quantum system and the external field. Condition (10) is not exact when one makes this calculation. The exact formula can be written by using formulae (80) of paper I. They are not given explicitly here because they present very little physical interest and it is much more useful to make a numerical calculation in each case. The correction is always very small. For $\delta = 0$ and $V_{12}/V_{13} = 10$ E_3 must be $2.9925 V_{12}$ instead of $3 V_{12}$. The supplementary effect of ν_{13} can be approximately deduced from formulae (80) by neglecting the nonresonant components of the internal field. This correction is of the order of

$$(13) \quad \delta \nu_{12} = \nu_{13}^2 / 8(V_{12} + |\delta|).$$

As $\delta \nu_{12} \ll \nu_{13}$ the approximation on the frequency of the reactant field is unimportant. Formula (12) implies also that the resonance width of level $|3\rangle$ is very small, of the order of $\frac{1}{4} A_{31}^{12}$. The greatest discrepancy between the qualitative model and the exact calculation lies in the neglect of the reaction of the field for the transition 1-2. In fact the component of the internal field at frequency $\Omega + 2\beta$ decays to zero with a decay rate $\underline{a}$, which is given, for a two-level atom, by STROUD and JAYNES⁽⁵⁾ in their formula (4.22). If $|\delta| \gg V_{12}$, $\underline{a} \rightarrow \frac{1}{2} A_{21}$. If $|\delta| \ll V_{12}$, the formula (4.22) can be approximated by

$$(14) \quad \underline{a} = (\delta/4 V_{12}) A_{21}.$$

Except in the case $\delta = 0$, where there is no damping, the reactant field at frequency $\Omega + 2\beta$ will decay at last. It will be a possibility for strongly populating the level $|3\rangle$ during a time τ_p of the order of $1/\underline{a}$ and only if at least one nutation can occur between levels $|1\rangle$ and $|3\rangle$ during the time τ_p . Condition (10) is necessary⁽¹²⁾ but not sufficient to allow a strong population of

⁽¹²⁾ All the approximations are valid for short times and then the resonant phenomenon will never start if condition (10) is not satisfied.

level $|3\rangle$. It must be completed by the condition

$$(15) \quad \nu_{13} > \underline{a},$$

or more explicitly, and in the limit $|\delta| < V_{12}$,

$$(16) \quad A_{31}^{12} > (\delta/V_{12})A_{21}.$$

But in all cases, except $\delta = 0$, the strong population of level $|3\rangle$ is a transient phenomenon which disappears after a time of the order of

$$(17) \quad \tau_p = (4V_{12}/\delta)/A_{21}.$$

For a given Δ it is the δ which satisfies the relation (10) with a minus sign which will give the more durable and important effect.

All these results have been checked by numerical integration of the system of differential equations of JAYNES and CRISP (⁴). No approximations have been made except the neglect of all frequency shifts. For all the calculations the parameters which have the dimension of a frequency are in units of V_{12} and the time is in units of $1/V_{12}$. We have displayed with a curve plotter $\rho_{33}(t)$ for some characteristic cases. For all the calculations $V_{13} = 0.1$ and $\varphi = 0$. In Fig. 2 we

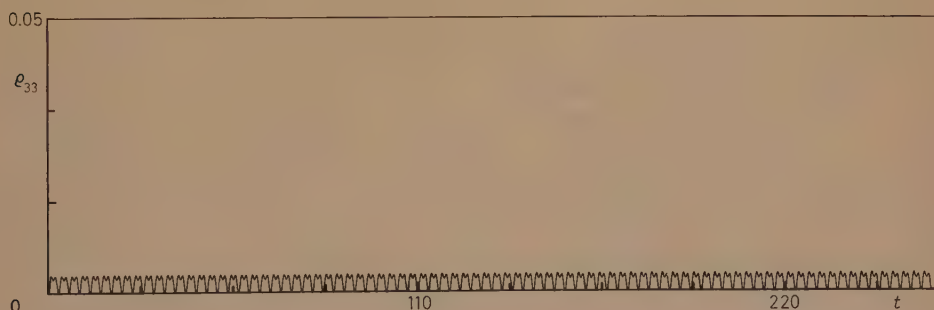


Fig. 2. - Population of the third level in the semi-classical model. $V_{13} = 0.1$, $A_{21} = 0$, $A_{31} = 0$, $\delta = 0$, $\Delta = 3$. In Fig. 2 to 6 and 13 V_{13} , A_{21} , A_{31} , δ , Δ are in units of V_{12} . Time is in units of V_{12}^{-1} .

have put $A_{21} = A_{31} = 0$, $\delta = 0$ and $\Delta = 3V_{12}$. $\rho_{33}(t)$ is always very small. With $A_{21} = 0.5$ and $A_{31} = 0.005$ it has been found that for $\delta = 0$ the resonance effect is maximum for $\Delta = 2.9935$ against 3.0 predicted by formula (10) and 2.9925 when taking into account the effect of V_{13} . We show in Fig. 3 $\rho_{33}(t)$ for $\Delta = 2.9935$ (curve A) and for a detuning frequency of level $|3\rangle$ from this position of 0.006 (curve B) and 0.012 (curve C). As this detuning corresponds to $\frac{1}{2}\nu_{13}$ and ν_{13} , this confirms our evaluation of the resonance bandwidth. In Fig. 4 the para-

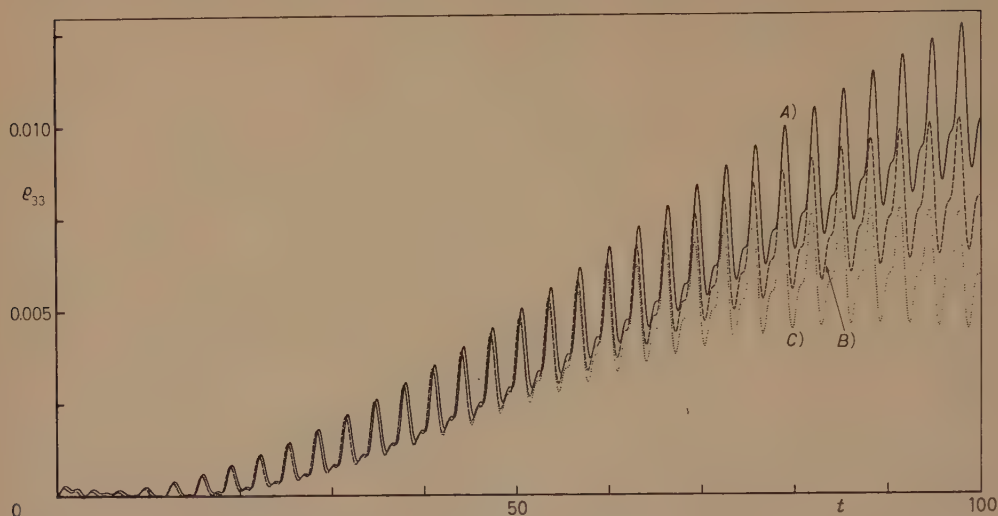


Fig. 3. - Population of the third level in the semi-classical model. $V_{13} = 0.1$, $A_{21} = 0.5$, $A_{31} = 0.005$, $\delta = 0$. In curve A) $\Delta = 2.9935$. For this value the resonance effect of the reactant field is maximum. In curve B) $\Delta = 2.9995$. In curve C) $\Delta = 3.0055$.

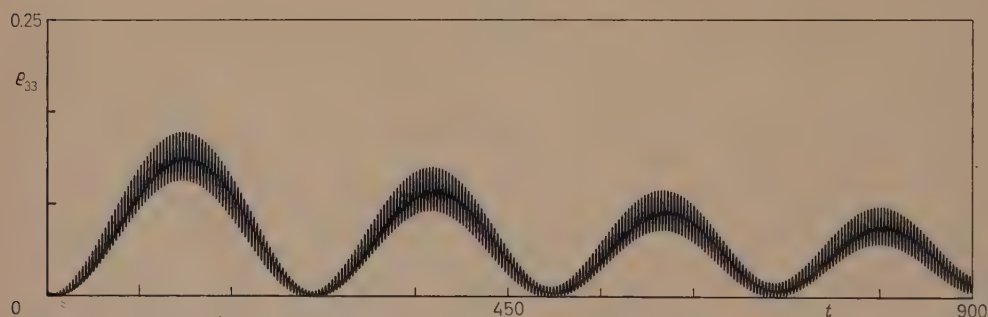


Fig. 4. - Population of the third level in the semi-classical model when the resonance effect is maximum. $V_{13} = 0.1$, $A_{21} = 0.5$, $A_{31} = 0.005$, $\delta = 0$, $\Delta = 2.9935$.

parameters are the same as for curve A) of Fig. 3, but the equations are integrated up to a time $t = 900$. The nutations between $|1\rangle$ and $|3\rangle$ are clearly visible. The period of the first nutation is 254, against our predicted result of 251 which was obtained with several drastic approximations. The periods of the following nutations are slightly decreasing, but remain in good agreement with our predictions. The small rapid oscillations are produced by the interaction between the matter and the components of the field at frequencies $\Omega - 2\beta$ and Ω . Figure 5 shows that, when the parameters satisfy the relation (10) with the plus sign, the population of level $|3\rangle$ reaches nearly the same value as it would if the feedback effect were not present. It is not a surprising result, as relation (15) is not satisfied with the parameters $\delta = 1.5$, $\Delta = 3$, $A_{21} = 0.5$, $A_{31} = 0.005$.

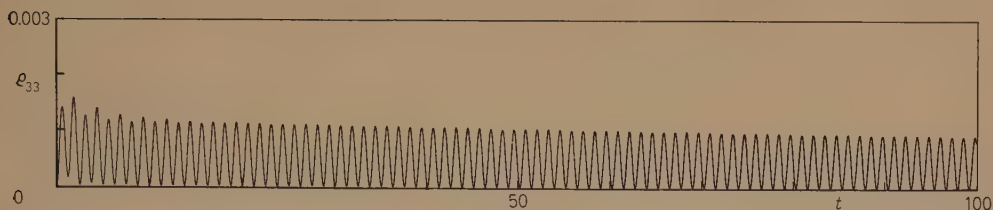


Fig. 5. — Population of the third level in the semi-classical model when condition (10) with the plus sign is satisfied. $V_{13} = 0.1$, $A_{21} = 0.5$, $A_{31} = 0.005$, $\delta = 1.5$, $\Delta = 3$.

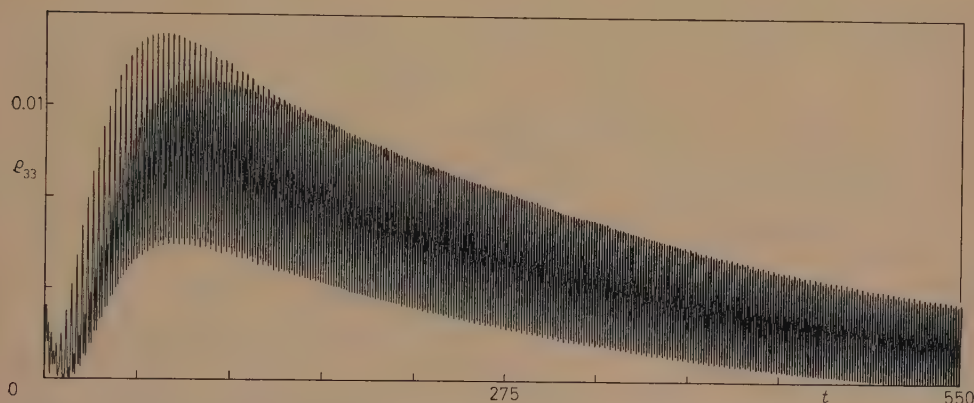


Fig. 6. — Population of the third level in the semi-classical model when condition (10) is satisfied and the frequency of the external field is not exactly the same as the frequency of the transition 1-2. $V_{13} = 0.1$, $A_{21} = 0.5$, $A_{31} = 0.005$, $\delta = 0.2$, $\Delta = 2.9$.

$\rho_{33}(t)$ does not exceed $0.16 \cdot 10^{-2}$, whereas in the similar case of Fig. 4 with $\delta = 0$ $\rho_{33}(t)$ reaches 0.148. But the parameters $\delta = 0$ can be used only if $\Delta \approx 3$. We show in Fig. 6 the behaviour of $\rho_{33}(t)$ for $\Delta = 2.9$, $A_{21} = 0.5$, $A_{31} = 0.005$. Relation (10) with a minus sign gives $\delta = 0.2$. The maximum value of $\rho_{33}(t)$ is 0.0126 against 0.148 in Fig. 4. Here again condition (15) is not satisfied.

3. — Quantum treatment.

In this Section we consider the same quantum system, but we treat the electromagnetic field and its interaction with the system quantum-mechanically. The external electromagnetic field is replaced by a mode k_0 of the field, containing N photons at time $t = 0$ and where N is a very great number. All the other modes are empty at time $t = 0$. They are treated as a reservoir, the interaction of which with the quantum system is responsible for the spontaneous emission. We use again the notations of Sect. 2 but for V_{12} and V_{13} which are replaced by $V_{12}^n = \alpha \sqrt{n} \mu_{12} \cdot e$ and $V_{13}^n = \alpha \sqrt{n} \mu_{13} \cdot e$. α is a coupling constant, n is

the number of photons in the $\mathbf{k}_0$ mode and $\mathbf{e}$ is the polarization vector of $\mathbf{k}_0$. The levels $|1\rangle$, $|2\rangle$ and $|3\rangle$ are replaced by levels $|1, n\rangle$, $|2, n\rangle$, $|3, n\rangle$ of the atom-plus-field system. An n -block of levels is set up with levels $|1, n\rangle$, $|2, n-1\rangle$, $|3, n-1\rangle$ which are closely grouped. We add to the atom-plus-field Hamiltonian the interaction Hamiltonian whose matrix elements are the V_{12}^n and only the matrix elements which connect nearly degenerate states are retained. (This is equivalent to the rotating-wave approximation in the semi-classical treatment.) If we call a_0 and $a_0^\dagger$ the annihilation and creation operators of the mode $\mathbf{k}_0$, and A_i and $A_i^\dagger$ the annihilation and creation operators of a state $|i\rangle$ of the quantum system, this Hamiltonian can be written

$$(18) \quad H_{12} = \alpha a_0 A_2^\dagger A_1 \boldsymbol{\mu}_{12} \cdot \mathbf{e} + \alpha a_0^\dagger A_1^\dagger A_2 \boldsymbol{\mu}_{12} \cdot \mathbf{e}.$$

H_{12} connects states of the same n -block. The eigenstates of the Hamiltonian $H_0 + H_{12}$ are again clustered into n -blocks and the eigenstates and energies for a block can be written ⁽³⁾

$$(19a) \quad |+, n\rangle = \sin \theta_n |1, n\rangle + \cos \theta_n |2, n-1\rangle,$$

$$(19b) \quad |-, n\rangle = \cos \theta_n |1, n\rangle - \sin \theta_n |2, n-1\rangle,$$

$$(19c) \quad |3, n\rangle,$$

$$(20a) \quad E_{+,n} = \frac{1}{2} \hbar (\delta + 2\beta_n + 2n\Omega),$$

$$(20b) \quad E_{-,n} = \frac{1}{2} \hbar (\delta - 2\beta_n + 2n\Omega),$$

$$(20c) \quad E_{3,n} = \hbar (\delta + \Delta + n\Omega),$$

with

$$(21) \quad \tan 2\theta_n = -2V_{12}^n/\delta$$

and

$$(22) \quad \beta_n = \frac{1}{2} (\delta^2 + 4V_{12}^{n*})^{\frac{1}{2}}.$$

These levels can be called the levels of the « *semi-dressed* » system. The system is not completely « *dressed* » ⁽¹³⁾ by the photons of the external field because the Hamiltonian does not contain the term which is responsible for the transition between levels $|1, n\rangle$ and $|3, n-1\rangle$ in each n -block. An n -block is populated from the $(n+1)$ -block by spontaneous emission. In another paper ⁽¹⁴⁾ we have treated a very similar problem but with a two-level system and $\delta = 0$. We have found that if at time $t = 0$ only the levels of block N

⁽¹³⁾ C. COHEN-TANNOUDJI and S. HAROCHE: *Compt. Rend.*, **262** B, 268 (1966).

⁽¹⁴⁾ G. OLIVER, E. RESSAYRE and A. TALLET: *Lett. Nuovo Cimento*, **2**, 777 (1971).

were populated, the levels of block n were reaching their maximum population at time $t = (N - n)/\Gamma_2$, where $\Gamma_2 = \frac{1}{2}A_{21}$. In our present problem the decay constant of level $|3\rangle$ is $\Gamma_3 = \frac{1}{2}A_{31}$. As it is also assumed that $A_{31} \ll A_{21}$, the cascade rate remains essentially governed by Γ_2 . As we have assumed that N is a very great number (of the order of 10^{17}) and that a typical value of Γ_2 is 10^8 Hz, we can assume that during the interaction the blocks which are involved in the cascade process will correspond with a very weak depopulation of the k_0 -mode. For a time of one hour the block which will reach its maximum population and will have an n of $0.9999964 \cdot 10^{17}$! For all the blocks of physical interest in our calculation we can replace n by N and V_{12}^n by $V_{12}^N = V_{12}$ and so on for θ and β . In principle the density matrix of the system has $9N^2$ elements. In terms of Lehmborg's formalism⁽¹⁵⁾ the differential-equation system which governs the time evolution of one element of the density matrix (in the basis of the naked atom $|1, n\rangle, |2, n\rangle, |3, n\rangle$) can be written, in a condensed form,

$$\begin{aligned}
 (23) \quad \dot{\rho}_{i,p,j,q} = & i(\Omega_j - \Omega_i + (q - p)\Omega)\rho_{i,p,j,q} + \\
 & + i \sum_{k>j} V_{k,j,q,q-1} \rho_{i,p,k,q-1} + i \sum_{k<j} V_{k,j,q,q+1} \rho_{i,p,k,q+1} - \\
 & - i \sum_{l<i} V_{i,l,p,p-1} \rho_{i,p-1,j,q} - i \sum_{l>i} V_{i,l,p,p+1} \rho_{i,p+1,j,q} - \\
 & - 2 \sum_{\substack{i<l \\ j<k}} \lambda^2 \rho_{i,p,k,q} \boldsymbol{\mu}_{il} \cdot \boldsymbol{\mu}_{kj} \Omega_{kj} \Omega_{il} |\Omega_{il}| + \sum_{\substack{k<j \\ k<l}} \lambda^2 \rho_{i,p,l,q} \boldsymbol{\mu}_{kj} \cdot \boldsymbol{\mu}_{lk} \Omega_{kj} \Omega_{lk} |\Omega_{lk}| + \\
 & + \sum_{\substack{i>l \\ k>l}} \lambda^2 \rho_{k,p,j,q} \boldsymbol{\mu}_{il} \cdot \boldsymbol{\mu}_{lk} \Omega_{il} \Omega_{lk} |\Omega_{lk}| ;
 \end{aligned}$$

i, j, k, l are states of the system and p and q are occupation numbers of the mode k_0 , $\Omega_{kj} = \Omega_k - \Omega_j$, λ is a coupling constant

$$(24) \quad \lambda = \left(\frac{2}{3} \hbar c^3\right)^{\frac{1}{2}}.$$

It is easy to see from eq. (23) that if $|i, p\rangle$ and $|j, q\rangle$ belong to the blocks r and s , they are coupled with elements of general form $\rho_{k,u,l,v}$ so that $|k, u\rangle$ and $|l, v\rangle$ belong respectively either to the r and s blocks or to the $r+1$ and $s+1$ blocks. On the other hand at time $t=0$ elements of the form $\rho_{i,p,j,q}$ vanish if $|i, p\rangle$ and $|j, q\rangle$ belong to different blocks.

This implies that we have only to calculate the elements of the density matrix between states belonging to the same block. We have replaced V_{12}^n and V_{13}^n by V_{12} and V_{13} . With this approximation eq. (23) is the same for any block except for a translation of numbers p and q . The relations be-

⁽¹⁵⁾ R. H. LEHMBERG: *Phys. Rev.*, **181**, 32 (1969).

tween p and q are always the same for a couple of subscripts i and j ($p = q$ if $i = j$, $p = q + 1$ if $i = 1$ and $j = 2$ or 3 and so on). Therefore we can have directly the reduced density matrix of the quantum system by summing over p from 0 to N . Obviously our approximations are not valid if $p \ll N$, but it makes no matter because for the time in which we are interested the blocks with such a value of p are practically unpopulated. This summation allows us to solve only a system of nine linear differential equations. They can be constructed on the basis of the eigenstates of the naked atom or of the «semi-dressed» atom. In eq. (23) the first term represents the natural evolution of the system. The four following terms correspond to the interaction between the system and the mode k_0 . The sixth term governs the population of a block by the levels of the next upper block. The last two terms govern two different effects. If $l = j$, they govern the spontaneous emission from the levels of a block to the levels of the immediately inferior block. If $l \neq j$, they show that a photon emitted from level $|j\rangle$ of block p is virtually absorbed by a suitable level $|k\rangle$ of block $p - 1$, immediately re-emitted, and absorbed by level $|l\rangle$ of block p . Therefore there is a transition between levels $|j\rangle$ and $|l\rangle$ of the same block through the levels of the immediately inferior block. In our particular case only levels $|2\rangle$ and $|3\rangle$ can be coupled. The coupling terms are $\lambda^2 \mu_{21} \cdot \mu_{13} \Omega_{21} \Omega_{13}^2$ or $\lambda^2 \mu_{31} \cdot \mu_{12} \Omega_{31} \Omega_{12}^2$. The ratio of these two terms is Ω_{31}/Ω_{21} . As it has been assumed that $\Delta \ll \Omega_2$, these two terms can be equated to a very good approximation.

We call the geometric mean of the two terms Γ_{23} and then we can write

$$(25) \quad \Gamma_{23} = (\Gamma_2 \Gamma_3)^{\frac{1}{2}} \cos \varphi$$

and replace the coupling terms by Γ_{23} .

All our preceeding results remain valid if the density matrix is written on the basis of the eigenstates of the «semi-dressed» system. In each block the state $|3\rangle$ is coupled with states $|+\rangle$ and $|-\rangle$ through the mode k_0 , the coupling constants being respectively $V_{13} \sin \theta$ and $V_{13} \cos \theta$. The state $|3\rangle$ is coupled with the states $|+\rangle$ and $|-\rangle$ through the spontaneous emission, the coupling constants being $\Gamma_{23} \cos \theta$ and $\Gamma_{23} \sin \theta$, respectively. In the same manner, as in the semi-classical treatment, δ is allowed to be negative, zero or positive and Δ is restricted to be positive. If we neglect the coupling with state $|-\rangle$, we can predict rigorously that the population of level $|3\rangle$ (*i.e.* $\varrho_{33}(t)$) will reach a maximum when $E_+ = E_3$. In fact, the interaction between states $|3\rangle$ and $|-\rangle$ will cause the maximum of ϱ_{33} to be reached for a value of E_3 slightly different from E_+ . Numerical evaluations of this shift have shown that it is much less than $V_{13} \sin \theta + \Gamma_{23} \cos \theta$ and the approximate relation $E_+ = E_3$ is sufficient to explain the condition for a strong population of level $|3\rangle$. From formulae (20) this condition can be rewritten as

$$(26) \quad \beta = \frac{1}{2} \delta + \Delta,$$

or

$$(27) \quad \delta = (V_{12}^2 - \Delta^2)/\Delta.$$

This condition is the same as condition (9) in the semi-classical treatment. The effects of the interaction with the mode $\mathbf{k}_0$ and of the spontaneous emission feedback cannot be distinguished except in the case where $V_{13} = 0$ (*i.e.* when the polarization vector of the mode $\mathbf{k}_0$ is orthogonal with the dipolar moment $\boldsymbol{\mu}_{13}$).

The differential-equation system which governs the time evolution of the density matrix is of the form

$$(28) \quad \frac{d\boldsymbol{\rho}(t)}{dt} = \mathbf{A}\boldsymbol{\rho}(t),$$

where $\boldsymbol{\rho}$ is a vector whose components are the $\rho_{ij}(t)$ and $\mathbf{A}$ is a non-Hermitian matrix. The system can be analytically solved by the method described in paper II. The solution is of the form

$$(29) \quad \rho_{ij}(t) = \sum_{k=1}^n c_{ij}^k \exp[\lambda_k t],$$

where the λ_k are the complex eigenvalues of $\mathbf{A}$. We define

$$(30) \quad \lambda_k = \gamma_k + i\omega_k.$$

At least one γ_k is zero and the others are negative. This is necessary for keeping the trace of the density matrix at 1. In our particular problem it has been found experimentally that only one λ_k is zero (we call it λ_1). c_{33}^1 is the value of $\rho_{33}(\infty)$. We call γ_2 the one of the γ 's which has the smallest nonzero absolute value. The value of $\rho_{33}(t)$ nearly reaches c_{33}^1 when $t > 1/\gamma_2$. The characteristic time τ_c is $1/\gamma_2$. $\rho_{33}(\tau_c)$ will have a nonnegligible value if condition (27) is satisfied and if the relation

$$(31) \quad V_{13} \sin \theta + \Gamma_{23} \cos \theta > 2\Gamma_3$$

is also satisfied. We have not been able to write the analytical expression for τ_c , but numerical calculations have shown that τ_c depends upon the ratio

$$(32) \quad r = (V_{13} \sin \theta + \Gamma_{23} \cos \theta)/\Gamma_2.$$

For $r \geq 1$ $\tau_c \approx 8/3\Gamma_2$. It reaches $1/2\Gamma_3$ when r reaches 0. If $r \geq 1$ the level $|3\rangle$ can be strongly populated by a cascade process even if relation (27) is not satisfied but the characteristic time τ_{caso} is much greater than τ_c . If τ_i is the

interaction time when the population of level $|3\rangle$ is measured, $\varrho_{33}(\tau_i, \Delta)$ (with all the other parameters unvarying) will exhibit a resonance at Δ_r determined by condition (27) only if $\tau_i \ll \tau_{\text{casc}}$. Otherwise $\varrho_{33}(\tau_i, \Delta)$ will be a function of Δ with a maximum at $\Delta_{\text{max}} < \Delta_r$ or even, if τ_i is sufficiently large, a decreasing function starting from 1 for $\Delta = 0$. The problem of the population of the two excited levels of the system in the general case will be treated elsewhere.

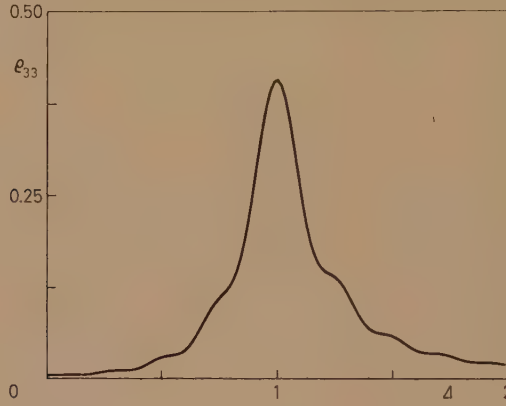


Fig. 7. — Population of the third level in the quantum theory at time $t = 3/\Gamma_2$. $V_{13} = 0.1$, $\Gamma_2 = 0.5$, $\Gamma_3 = 0.005$, $\Gamma_{23} = 0.05$, $\delta = 0$. In Fig. 7 to 12 and 14 V_{13} , Γ_2 , Γ_3 , Γ_{23} , δ and Δ are in units of V_{12} .

In Fig. 7 to 12 we display $\varrho_{33}(\tau_i, \Delta)$ against Δ with $\tau_i \approx \tau_c$ and for various sets of parameters. All the quantities which have the dimension of a frequency are in units of V_{12} and the time is in units of $1/\Gamma_2$. For each value of δ we study two configurations, the first with the two dipolar moments and the polarization vector of $\mathbf{k}_0$ all parallel (Fig. 7, 9 and 11) and the second with the two dipolar

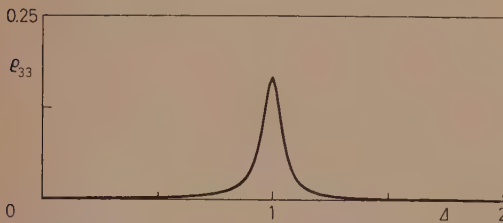


Fig. 8.

Fig. 8. — Population of the third level in the quantum theory at time $t = 50/\Gamma_2$. $V_{13} = 0$, $\Gamma_2 = 0.5$, $\Gamma_3 = 0.005$, $\Gamma_{23} = 0.03535$, $\delta = 0$.

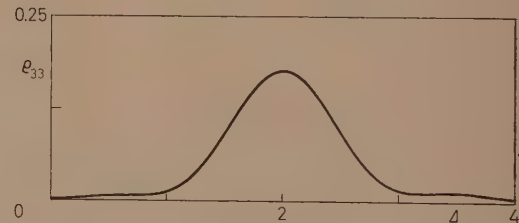


Fig. 9.

Fig. 9. — Population of the third level in the quantum theory at time $t = 3/\Gamma_2$. $V_{13} = 0.1$, $\Gamma_2 = 0.5$, $\Gamma_3 = 0.005$, $\Gamma_{23} = 0.05$, $\delta = -1.5$.

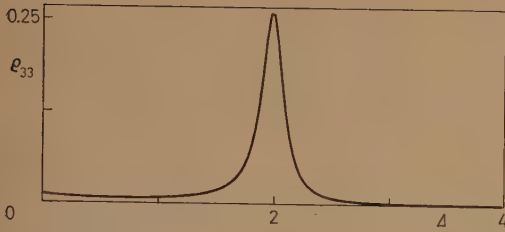


Fig. 10.

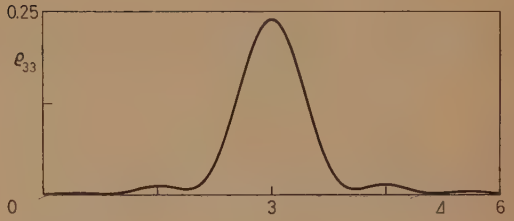


Fig. 11.

Fig. 10. - Population of the third level in the quantum theory at time $t = 50/\Gamma_2$. $V_{13} = 0$, $\Gamma_2 = 0.5$, $\Gamma_3 = 0.005$, $\Gamma_{23} = 0.03535$, $\delta = -1.5$.

Fig. 11. - Population of the third level in the quantum theory at time $t = 3/\Gamma_2$. $V_{13} = 0.1$, $\Gamma_2 = 0.5$, $\Gamma_3 = 0.005$, $\Gamma_{23} = 0.05$, $\delta = -2.6667$.

moments angled at 45° and the polarization vector orthogonal to μ_{13} , *i.e.* $V_{13} = 0$ (Fig. 8, 10, 12). δ has been chosen to give the resonant effect for $\Delta = V_{12}$ (Fig. 7 and 8), $\Delta = 2V_{12}$ (Fig. 9 and 10) and $\Delta = 3V_{12}$ (Fig. 11 and 12). Relation (27) is very well verified in all the cases.

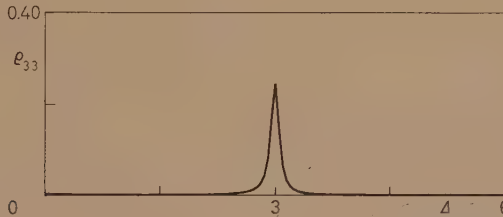


Fig. 12. - Population of the third level in the quantum theory at time $t = 50/\Gamma_2$. $V_{13} = 0$, $\Gamma_2 = 0.5$, $\Gamma_3 = 0.005$, $\Gamma_{23} = 0.03535$, $\delta = -2.6667$.

4. - Discussion.

When studying the effects of the interaction between electromagnetic field and matter, the predictions of semi-classical and quantum theories are never exactly the same, but in most cases ^(3,6) experiments would hardly decide between them. Recently it has been pointed out that there are at least two experiments where the semi-classical theory does not predict correctly the characteristics of the electromagnetic field emitted by the matter ^(2,7). In this work we have found a case where the two theories give the same condition for a strong population of the third level and another case where only the semi-classical theory predicts a strong population of the third level when a particular condition is satisfied. The first case presents a physical interest because it predicts an anomalous population of a level which would not be strongly pop-

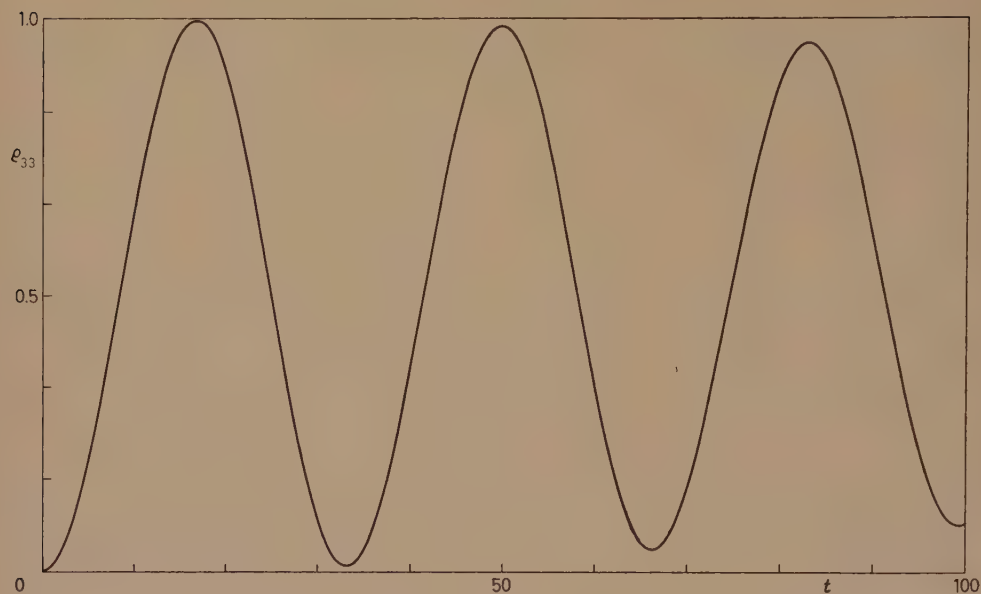


Fig. 13. - Population of the third level in the semi-classical model when condition (9) is satisfied. $V_{13} = 0.1$, $A_{21} = 0.5$, $A_{31} = 0.005$, $\delta = -2.6667$, $\Delta = 2.9925$.

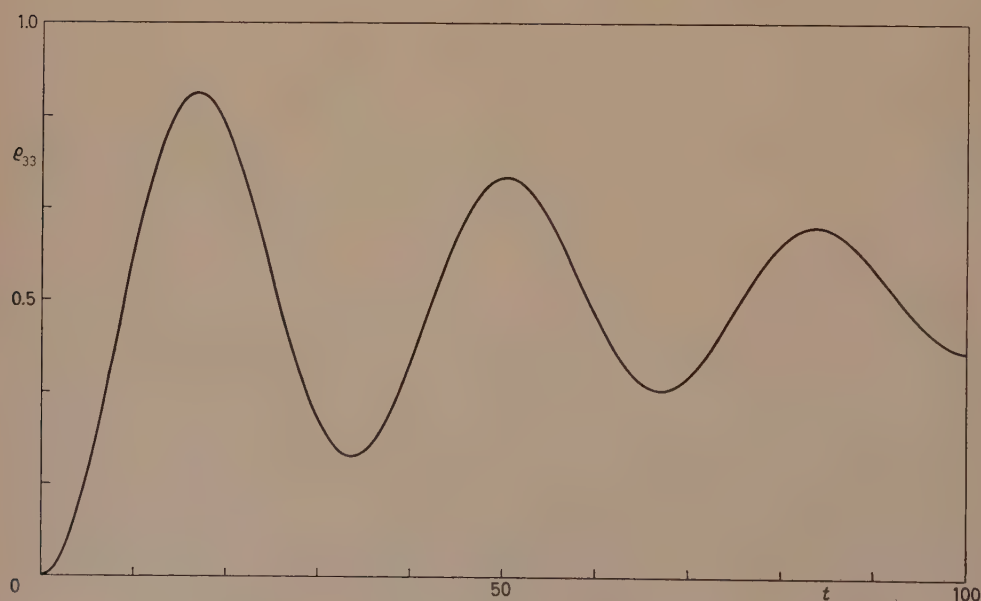


Fig. 14. - Population of the third level in the quantum theory when condition (27) is satisfied with the same parameters as in Fig. 13. $V_{13} = 0.1$, $\Gamma_2 = 0.25$, $\Gamma_3 = 0.0025$, $\Gamma_{23} = 0.025$, $\delta = -2.6667$, $\Delta = 2.9925$. t is in units of V_{12}^{-1} .

ulated according to Bohr's rule. We show in Fig. 13 and 14 $\varrho_{33}(t)$ calculated with the semi-classical theory (Fig. 13) and with the quantum theory (Fig. 14) when condition (9) or (27) is satisfied. Although the behaviour of $\varrho_{33}(t)$ is not exactly the same in the two approaches, one cannot be sure that an experiment would decide between the two theories. On the contrary, when condition (10) is satisfied, the semi-classical theory predicts a strong population of level $|3\rangle$, whereas the quantum theory predicts a very weak population. In this case it will be much easier to make an experiment which would decide between the two theories in a case where they disagree about the behaviour of the quantum system and not only of the emitted light ^(2,7).

The quantum treatment assumes that the system is driven by a strongly populated mode, whereas in the semi-classical treatment the system is driven by a classical electric field. The quantum equivalent of our classical field is a Glauber field ⁽¹⁶⁾ which is defined by

$$(33) \quad |\Phi_{\bar{n}}\rangle = \exp\left[-\frac{1}{2}\bar{n}\right] \sum_{n=0}^{\infty} \bar{n}^n (n!)^{-\frac{1}{2}} |n\rangle,$$

where n is an occupation number of the mode k_0 and $\bar{n}$ is a mean number of photons defined by

$$(34) \quad V_{12}^{\bar{n}} = V_{12}^{\text{semi-classical}}.$$

For a strong field it can be assumed that the $\bar{n}$ which satisfies relation (34) is an integer number. If w_0 is the weight of state $|\bar{n}\rangle$ in $|\Phi_{\bar{n}}\rangle$, the weight of state $\bar{n} \pm p$ is approximately

$$(35) \quad w_p = w_0 \exp[-p^2/2\bar{n}],$$

e.g. $w_p = 10^{-6}w_0$ if $p = 5.256\bar{n}^{\frac{1}{2}}$. At time $t=0$ for all n corresponding to an appreciably populated block V_{12}^n can be equated to $V_{12}^{\bar{n}}$ to a very good approximation because $\bar{n}$ is a very great number. The rate of the cascade process is not affected by the fact that many blocks are populated at time $t=0$. The hypothesis of Sect. 3, that all the blocks involved in the cascade process during the time of the interaction have nearly the same V_{12}^n remains valid. If we are only interested in the populations of the levels of the quantum system, we can always sum over the n and we have again to solve the same system of nine equations as before and with the same initial condition $\varrho_{11}(0) = 1$. The populations of the levels of the system are the same if the system is driven by a strong Glauber field or by a strongly populated mode. The principal effect

⁽¹⁶⁾ R. J. GLAUBER: *Phys. Rev.*, **131**, 2768 (1963).

of the replacement of the mode by the Glauber field is to induce a polarization of the system, but this is unimportant for our problem. Formula (35) shows that the population of the block $9\bar{n}$ (which corresponds to $V_{12}^{9\bar{n}} = 3V_{12}^{\bar{n}}$) is nearly zero and no effect of population of level $|3\rangle$ with $E_3 \approx 3V_{12}^{\bar{n}}$ can arise.

The quantum treatment using Lehmborg's formalism does not give the same results as the semi-classical one because it does not permit the following process: emission from level $|+, n\rangle$ to level $|-, n-1\rangle$ of a photon at frequency $\Omega + 2\beta$, emission from level $|-, n-1\rangle$ to level $|+, n-2\rangle$ of a photon at frequency $\Omega - 2\beta$, and absorption from level $|+, n-2\rangle$ to level $|3, n-1\rangle$ of the photon precedently emitted with frequency $\Omega + 2\beta$. HAAKE⁽¹⁷⁾ has shown that, in the time-ordered series of DYSON, all terms of the form $a_k^\dagger(t_1) a_l^\dagger(t_2) a_k(t_3)$ (and more generally all terms where $a_k^\dagger(t_i)$ and $a_k(t_j)$ are separated by one or more field operators of the other modes) give a negligible contribution to the density matrix of the system if the density of modes $\varrho(|\mathbf{k}|)$ is a sufficiently «soft» function. As this is obviously our case, we can say that the use of Lehmborg's formalism is not responsible for the discrepancy between the semi-classical and quantum treatments.

The best method to test the validity of the two theories seems to irradiate a three-level quantum system with a pulse of monochromatic light of suitable frequency and, after the pulse has been switched off, to study the light spontaneously emitted during the transition from the level $|3\rangle$ to the fundamental $|1\rangle$. When condition (9) or (27) or condition (10) is satisfied, the quantity of light emitted at frequency Ω_3 must be much more important than in the other circumstances. Practically it is only in an atom or, possibly, in a diatomic molecule, that one can really isolate a three-level system. To avoid the problem of the damping of the spontaneous emission the frequency of the external field Ω must be as close as possible to Ω_2 . It is unimportant from the theoretical point of view if Ω_3 is varying with V_{12} constant or if V_{12} is varying with Ω_3 constant, but it seems that the second method would be more practicable. Formula (12) predicts that, if condition (10) is satisfied, $\varrho_{33}(t)$ will reach its first maximum at $t \geq 2\pi/A_{12}^{13}$ and numerical calculation show that this result is roughly correct. A pulse duration τ_p of the order of $2\pi/A_{21}^{13}$ is sufficient. If $(V_{13} + \frac{1}{2}A_{21}^{13})/\sqrt{2} < \frac{1}{2}A_{21}$ there is no disadvantage to have $\tau_p > 2\pi/A_{21}^{13}$, but τ_p will have to be fitted in such a manner that the system will be in the excited state at the end of the pulse. If $(V_{13} + \frac{1}{2}A_{21}^{13})/\sqrt{2} > \frac{1}{2}A_{21}$, numerical calculations in the quantum case show that cascade processes can give rise to a nonnegligible population of the level $|3\rangle$ for a time $t \geq \tau'_{\text{casc}}$. τ'_{casc} has not the same values as τ_{casc} defined in Sect. 3 because τ_{casc} corresponds to $\Delta \approx 3V_{12}$, whereas τ'_{casc} corresponds to $\Delta \approx V_{12}$ (with $\delta \approx 0$ in the two cases). Numerical calculations show that, even for $V_{13} \gg \Gamma_2$, τ'_{casc} is of the order of a fraction of $1/\Gamma_3$.

(17) F. HAAKE: *Zeits. Phys.*, **223**, 353 (1969).

It is necessary to ensure that $\tau_p \ll \tau'_{\text{casc}}$. But the most satisfactory method to avoid all these difficulties is to make $V_{13} < A_{21}$. The ideal condition is $V_{13} = 0$. It will be attained if the angle φ between μ_{12} and μ_{13} is neither 0° nor 90° and if all the atoms or molecules of the sample have their dipolar moments oriented in the same directions so that, with a conveniently polarized light, one will have $V_{13} = 0$ but $A_{21}^{13} \neq 0$. In any case, if the population of ϱ_{33} is due to the spontaneous-emission feedback, it will be a resonant effect; if it is due to the cascade process it will be a nonresonant effect. If there is a damping process other than the spontaneous emission, *e.g.* a collision damping, the homogeneous broadening corresponding to this process must be at most of the order of Γ_3 . It is also essential that the inhomogeneous broadenings of the two excited states (*e.g.* Doppler effect broadening) do not overlap. It was not an easy task to find a three-level system which satisfies all our conditions. Our calculation implies that all the three-level systems of the sample have the same orientation. Therefore molecules can be used only if they are trapped in a suitable matrix with only one site. On the contrary the experiment can be made with atomic vapour because of the spherical symmetry of the atoms. Several atoms give suitable three-level systems, but for most of them (S, Cl, Br, I, Os) the present laser technology does not allow one to satisfy the conditions on ω_{21} or on V_{12} . The only practicable atom is Y (¹⁸). State $|1\rangle$ corresponds to the fundamental level $^2D_{3/2}$, state $|2\rangle$ to the level $^2F_{5/2}$ and state $|3\rangle$ to level $^4F_{3/2}$, $\omega_{21} = 15326.83 \text{ cm}^{-1}$ and $\omega_{31} = 15329.04 \text{ cm}^{-1}$, $\delta = 2.21 \text{ cm}^{-1}$. The required V_{12} is 0.74 cm^{-1} . All these parameters are compatible with the existing dye-lasers.

All our discussion remains valid for observing the anomalous population of level $|3\rangle$ when condition (9) or (27) is satisfied. Figure 13 (semi-classical case) and 14 (quantum case) show that, for the same parameters, the behaviour of $\varrho_{33}(t)$ is nearly the same in the two theories. In the quantum approach level $|3\rangle$ can be strongly populated, even if condition (27) is not satisfied, if $\tau_p > \tau_{\text{casc}}$ (defined in Sect. 3). In this case the population effect is not selective. Practically it seems difficult to test the validities of the two theories in a case where condition (9) or (27) is satisfied. This anomalous population effect remains still interesting by the fact that it reveals how the levels of the system are shifted under the effect of the external field.

It is interesting to point out that the discrepancy between the two theories has its origin in the following circumstances. In the semi-classical theory any component $E_{ij}(t)$ of the reactant field (corresponding to the component $p_{ij}(t)$ of the polarization) can induce transitions between any two pseudolevels $|k\rangle$ and $|l\rangle$ of the system if $|\omega_{ij}| \approx |\omega_{kl}|$, whereas, in the quantum theory, population is transmitted from a level of the dressed system to another by a two-

(¹⁸) C. E. MOORE: *Atomic Energy Levels*, Vol. 2 (Washington, 1952).

photon process which implies that the two levels have nearly the same energy. This fundamental difference remains in all the semi-classical theories obtained by coupling Schrödinger equation with Maxwell equations. These theories have worked satisfactorily in many cases, particularly for superradiant photon echoes⁽¹⁹⁾ and self-induced transparency⁽²⁰⁾ but always with two-level systems. Similar experiments with three-level or many-level systems may give different conclusions.

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One of us (G.O.) would like to thank Dr. A. TALLET for many helpful discussions.

(¹⁹) A. M. PONTE GONÇALVES, A. TALLET and R. LEFEBVRE: *Phys. Rev.*, **188**, 576 (1969); A. COMPAAN and I. D. ABELLA: *Phys. Rev. Lett.*, **27**, 23 (1971).

(²⁰) S. L. MC CALL and E. L. HAHN: *Phys. Rev. Lett.*, **18**, 908 (1967); *Phys. Rev.*, **183**, 457 (1969); C. K. N. PATEL and R. E. SLUSHER: *Phys. Rev. Lett.*, **19**, 1019 (1967); C. K. N. PATEL: *Phys. Rev. A*, **1**, 979 (1970); A. M. PONTE GONÇALVES, A. TALLET and R. LEFEBVRE: *Phys. Rev. A*, **1**, 1472 (1970).

● RIASSUNTO (*)

Si studia il comportamento di un sistema quantico a tre livelli con uno stato fondamentale e due stati eccitati raggruppati strettamente durante la sua interazione con un campo elettromagnetico monocromatico forte. Il c.e.m. accoppia fortemente ed in modo risonante il livello fondamentale (primo livello) con un livello eccitato (secondo livello) ed accoppia debolmente ed in modo non risonante il livello fondamentale con l'altro livello eccitato (terzo livello). Si usano successivamente la teoria semiclassica della radiazione di Jaynes e Crisp e la teoria quantistica della radiazione, nel formalismo di Lehmeberg, per calcolare la matrice di densità del sistema. Si trova che, per parametri dati che caratterizzano il campo elettromagnetico ed i primi due livelli, ci sono due particolari dell'intervallo di frequenza fra i livelli eccitati per cui il terzo livello può essere densamente popolato. Per il primo valore le due teorie danno risultati consistenti ed il terzo livello è popolato sotto l'effetto sia del campo esterno che del campo emesso spontaneamente dal sistema (campo di reattanza o interno). Per il secondo valore, solo la teoria semiclassica predice una densa popolazione per il terzo livello, che è popolato solo per effetto del campo interno. Si predicono approssimativamente i valori dell'intervallo di frequenza usando i concetti di « pseudolivello » nel campo semiclassico e di sistema « semivestito » nel caso quantistico. Si propone di usare la discrepanza fra le due teorie nel secondo caso per controllare sperimentalmente la validità delle teorie semiclassica e quantistica della radiazione.

(*) Traduzione a cura della Redazione.

Обратная связь по спонтанному излучению в трех-уровневой квантовой системе: Случай противоречия между квазиклассической и квантовой теориями излучения.

Резюме (*). — Исследуется поведение трех-уровневой квантовой системы, которая имеет основное состояние и два близкорасположенных возбужденных состояния, при взаимодействии с сильным монохроматическим полем. Электромагнитное поле связывает сильно и резонансным образом основной уровень (первый уровень) с одним из возбужденных уровней (второй уровень), и связывает слабо и нерезонансным образом основной уровень с другим возбужденным уровнем (третий уровень). Квазиклассическая теория излучения Джейнса и Криспа и квантовая теория излучения, в формализме Лемберга, последовательно используются для вычисления матрицы плотности рассматриваемой системы. Получается, что для заданных параметров, характеризующих электромагнитное поле и первые два уровня, существуют два значения частотного интервала между возбужденными уровнями, для которых третий уровень может быть сильно заселен. Для первой величины эти две теории дают согласующиеся результаты и третий уровень заселяется и под действием внешнего поля и под действием поля, спонтанно излучаемого системой (реагирующее или внутреннее поле). Для второй величины только квазиклассическая теория предсказывает сильную заселенность для третьего уровня, причем, заселение происходит под действием только внутреннего поля. Указанные значения частотного интервала приблизительно предсказываются с помощью концепции «псевдо-уровней» в квазиклассическом случае и концепции «квази-одетой» системы в квантовом случае. Предсказания уточняются благодаря численным расчетам. Предлагается использовать различие между двумя теориями во втором случае, чтобы экспериментально проверить справедливость квазиклассической и квантовой теорий излучения.

(*) *Переведено редакцией.*

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11 Marzo 1973

Il Nuovo Cimento

Serie 11, Vol. 14 B, pag. 104-126

